REMARKS

Amendments to the Claims:

Applicants respectfully request that the following amendments be entered.

Claim 1 has been amended to incorporate the limitation of original claim 10 into new claim 1, with some modifications. New claim 1 now provides in step (d) that the fuel cell is heated to an intermediate temperature between the initial temperature and the desired temperature while the fuel cell is operated in an open circuit state. New step (c) of claim 1 provides that the fuel cell is then operated at a closed circuit state (an external circuit is connected to the fuel cell) **before** the intermediate temperature equals the desired temperature. Thus, in the method of new claim 1, the fuel cell is heated to a temperature below the desired temperature by operating it in a closed state, and the remaining heating up to the desired temperature is done while in the closed circuit state. The advantage of this is that the heating of the fuel cell is accelerated when it is switched to a closed circuit state (see paragraph 33 of the disclosure).

Support for this amendment to new claim 1 may be found in paragraphs 27 and 33 of the disclosure.

New claims 10 and 20 have each been amended to specify that the initial temperature is below the freezing point of water and the intermediate temperature is above the freezing point of water. Support for these amendments may also be found in paragraphs 27 and 33 of the disclosure.

Claim Rejections – 35 USC 102(a):

In the Office Action, the Examiner has rejected pending claims 1-20 as being "clearly anticipated" by WO 01/52339. Attached to this response is a machine translation of WO 01/52339 since the original is in German. Applicants submit that new claim 1 is now not clearly anticipated by WO 01/52339.

WO 01/52339 does not mention in explicit terms that the fuel cell is heated by operating it in an open circuit state. At most, WO 01/52339 only says that temperature monitoring is done "with a switched-off fuel cell system" (page 5, middle paragraph of the translation). There is no other

mention or suggestion in WO 01/52339 that the fuel cell operates in an open circuit state while it is heating up.

Moreover, WO 01/52339 clearly does not teach or suggest steps (d) and (e) in the method of new claim 1. If in fact WO 01/52339 does disclose operation of the fuel cell in an open state (although this is debatable), it clearly fails to teach heating the fuel cell to an intermediate temperature below the desired temperature and then operating the fuel cell in a closed circuit state until the desired temperature is reached. The advantage to this series of steps is that the heating of the fuel cell is accelerated (see paragraph 33 of the disclosure). Therefore, steps (d) and (e) in new claim 1 now distinguish the invention claimed from the method disclosed in WO 01/52339.

Claim Rejections – 35 USC 102(a):

In the Office Action, the Examiner has also rejected pending claims 1-20 as being anticipated by Colbow. The Examiner noted that paragraph 53 of Colbow teaches that the fuel cell can be heated while operating in an open circuit state.

Colbow does not, however, teach or suggest the method of new claim 1. In particular, Colbow fails to disclose heating the fuel cell to an intermediate temperature below the desired temperature and then operating the fuel cell in a closed circuit state until the desired temperature is reached. Colbow is therefore similar to WO 01/52339 in this respect, that is, neither of the two references suggest these additional steps and the advantage that the heating of the fuel cell is accelerated. Therefore, steps (d) and (e) in new claim 1 now also distinguish the invention claimed from the method disclosed in Colbow.

For all these reasons, Applicants respectfully submit that new claim 1 as amended, and dependent claims 2-20 distinguish over the teachings of WO 01/52339 and Colbow.

A Petition for an Extension of Time requesting an extension of one month for filing the subject response is enclosed. The Commissioner is authorized to charge any deficiency or credit any overpayment in the fees for same to our Deposit Account No. 500663. A signed copy of this page is enclosed if required for this purpose.

In view of the foregoing, allowance of the above-referenced application is respectfully requested.

Respectfully submitted,

DINO P. CLARIZIO
Attorney for Applicant
Registration No. 37,572
Telephone: 416-971-7202
Facsimile: 416-971-6638

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(71) Applicant *(for all the States indicated except US)* DAIMLER CHRYSLER AG [DE/DE]; Epplestrasse 225, 70567, Stuttgart (Germany)

(72) Inventors; and

(75) Inventors/Applicants *(for US only)* Müller, Jens Thomas [DE/DE]; Fünf-Bäume-Weg 195, 89081 Ulm (Germany)

(74) Agent: DAHMEN, Toni et al., Daimler Chrysler AG, Intellectual Property Management, FTP – C 106, 70546 Stuttgart (Germany)

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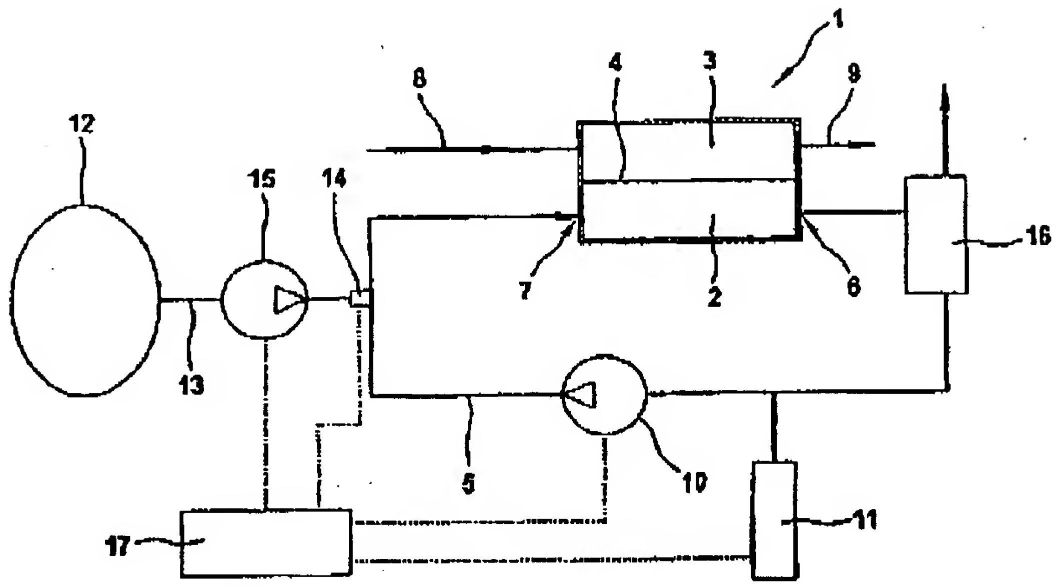
(54) Title: Liquid-fuel cell system

(54) Title: LIQUID-FUEL-CELL SYSTEM

(54) Bezeichnung: FLÜSSIG BRENNSTOFFZELLENSYSTEM



(57) Abstract: The invention relates to a fuel cell system comprising an anode chamber (2) and a cathode chamber (3), which are separated from one another by a proton-conductive membrane (4). A gas containing oxygen flows through the cathode chamber. A liquid fuel/coolant mixture, preferably a mixture of methanol and water is guided in a circuit through the anode chamber. To provide greater frost-protection and improved cold-starting capability, a temperature in the fuel cell system is monitored, even when said system is at a standstill and if a drop in temperature occurs, the concentration of fuel in the anode circuit is increased.

The invention concerns a fuel cell system and a technique for driving such a fuel cell system with the features in accordance with the general terms of Patent Claim 1 and/or 6.

For the daily-use service life capability of fuel cell systems, in particular with regard to application in vehicles, frost resistance and cold-weather starting capability are essential criteria for the fuel cell systems because of the amounts of water involved, which is a problem. Also, for the so-called direct methanol fuel cell (DMFC) which, because of direct operation with a liquid fuel medium, very extensive system simplifications are needed, so that so far the cold-weather starting problems are still unresolved.

A measuring type of fuel cell system is well-known from DE 198 07 876 A1. Here, on the anode side a liquid methanol/water mixture flows in the circuit. To monitor the performance using a constant methanol concentration from a supply vessel, methanol is metered while flowing into the anode circuit. The metering amount is thereby determined with the help of a concentration sensor in the running anode circuit. As coolants, liquids or ionic and/or nonionic additives used with water, with good freeze protection characteristics, are suggested.

In particular for a DMFC, such suitable coolants are not known at present and probably will not be available in the foreseeable future. The technological background is as follows:

The DMFC is usually processed at temperatures around approximately 100 C. The appropriate methanol concentration is typically between 0.5 and 2 mol/l and/or a weight percentage of 1.6 and 6.4. One cause is the methanol permeability of the available membranes. If methanol is used in higher concentrations, the residual methanol diffuses by way of the membrane to the cathode. The consequence is a drastically reduced efficiency. On the other hand, the cryoscopic constant of the water is 1.86 K kg/mol, which signifies the mol/kg of additive whose freezing point drops only 1.86 C. With

regard to colligative characteristics, this value is independent of the type of additive. The freezing point of the ordinarily used water/methanol mixtures thereby lies at approximately -1 to -4 C.

In order to ensure, for example, freeze protection to -30 C, however, an additive in a concentration of over 16 mol/kg is required. Such an additive is currently not available. It is not expected that it will be available in the near future, because even a relatively small molecule with one assumed mol mass of 50 g/mol is required at a concentration of 800 g/kg. A mixture of this composition is not available to be supplied to the anode in a stoichiometric amount with water. For the anode reaction, however, water and methanol in the stoichiometric relationship of 1:1 is required. All salts, acids, and bases are not applicable as freeze protection additives, because they lead to an increased electrical conductivity of the cooling water and thus inevitably lead to short-circuit currents in the apparatus.

The function of the invention is to offer a fuel cell system driven by means of a liquid fuel medium/coolant mixture and a technique for driving such a fuel cell system with improved frost protection and cold-weather starting characteristics.

The function is solved according to the invention by the characteristic features of the Patent Claim 1 and/or 6.

By an increase in the fuel medium concentration in the anode circuit line at decreasing temperatures, the freezing point of the fuel medium/coolant mixture is increased, thus allowing freeze protection, whereby the efficiency in the normal operation of the system does not worsen at the same time. With these preventative measures, freeze protection to -35 C is possible. At the same time, the cold-start condition is improved by a faster temperature rise of the fuel cell, because the fuel medium is diffused due to the higher concentration increased by the membrane to the cathode, and oxidized immediately there after the start of the air supply catalytically under heat. Thus, the cold-weather starting procedure is substantially accelerated.

Components can be saved and thus the costs and required building space can be reduced by the use of a combined concentration and temperature sensor.

The frost resistance can be ensured in a simple manner, in which the higher fuel-medium concentration provided in the anode circuit line is adjusted either by continuous adaptation of the theoretical concentration value to the decreasing temperature, or is adjusted by comparison of the determined temperature with one temperature threshold precipitously.

The additional required fuel-medium amount can, despite sufficient freeze protection, be reduced, so the efficiency altogether can be improved by the application of several temperature thresholds. In this case, namely the system is not always changed over immediately when falling below a temperature threshold value for maximum freeze protection, but the freeze protection is adapted to the actual temperature.

By the activation of the temperature monitoring only with a switched-off fuel cell system, the [energy] expenditure is reduced. At the same time, however, there are no disadvantages, because the system is always sufficiently hot during operation and therefore no additional freeze protection is necessary.

Other benefits and arrangements are evident from the subclaims and the detailed description. The invention is described in more detail using a drawing, which simplifies the principle construction of the represented fuel cell system.

Overall, 1 indicates the fuel cell consisting of an anode chamber 2 and a cathode chamber 3, which are separated from each other by a proton-conducting membrane 4. Over an anode circuit line 5, which connects the anode chamber outlet 6 with an anode chamber inlet 7 of the fuel cell 1, a liquid fuel medium/coolant mixture flows by the anode chamber 2. As the fuel medium here, suitable substances that are electrochemically oxidizable at room temperature can be used. The system described in the application

example is driven with liquid methanol as the fuel medium and with water as a coolant. Although in the following only the use of a methanol/water mixture is described, the protected range of this patent registration is not to be restricted to this application example. Such a system driven with a liquid methanol/water mixture becomes is usually described as a direct methanol fuel cell (DMFC).

Into cathode chamber 3 over a cathode inlet 8 an oxygen-containing gas is conducted. In accordance with application example, ambient air is used. In the fuel cell 1 the fuel medium at the anode is oxidized, which is reduced by atmospheric oxygen at the cathode. For this, the proton-conducting membrane 4 is coated on the suitable surface arc with suitable catalysts, for example, higher surface area noble-metal or catalysts supports. From the anode side, protons can now move by the proton-conducting membrane 4 and interconnect to the cathode side with the oxygen ions to form water. With this electro-chemical reaction, a voltage results between the two electrodes. By parallel and/or tandem arrangement of many of such cells to form a so-called stack arrangement, higher voltages and a strong current can be achieved.

As the product, carbon dioxide gas enriched with water and methanol forms at the anode outlet. This liquid/gas mixture is passed over the anode circuit line 5 from the anode chamber 2. The remaining oxygen and water vapor containing exhaust air flow from the cathode over a cathode exit fuel line 9. A good efficiency can be obtained in the cathode chamber 3 preferably prepared with high-pressure ambient air.

On the anode side, the methanol/water mixture is circulated with the help of a pump 10 with a given pressure through the anode circuit line 5. The ratio of water to methanol in the anode circuit line 5 is adjusted with the help of a sensor 11, which measures the methanol concentration in the anode circuit line 5. As a function of this sensor signal, then usually concentration control of the methanol/water mixture takes place, whereby liquid methanol from a methanol supply vessel 12 over a supply line 13 is tension force-driven and is not injected with the help of fuel injector 14 into the anode circuit line 5. The injection pressure is created by an injection pump 15 arranged in the supply line 13.

The methanol dosage takes place via a suitable triggering of the fuel injector14. For this, use is made of controller 17, which is regulated by measuring and/or control lines connected with the pump 10, sensor 11, injection pump 15, fuel injector 14, and other components, if necessary. In the anode chamber 2, a methanol/water mixture with a preferably more constant concentration is tension force-driven. In addition, it is conceivable to also vary the methanol concentration during the operation of the fuel cell system.

On the anode side, with the help of a gas separator 16, carbon dioxide enriched with methanol and water vapor is discharged from the liquid/gas mixture in the anode circuit line 5. An excessively high methanol discharge over the carbon dioxide gas is to be prevented, since otherwise the overall efficiency of the system is reduced and, at the same time, noncombusted methanol is not discharged into the environment. In the drawing, the represented gas separator is a simpler version of the more complex device used.

Further, a device is used for the determination of the intended temperature T_{actual}. For this, common temperature sensors can be used. Advantageously, the sensor 11 is used as a combined concentration and temperature sensor. Thus, additional components are not necessary. It is, however, obvious that a separate temperature sensor can be used. According to an application example, the sensor 11 is arranged in the anode circuit line 5 between the gas separator 16 and the pump 10. It is however also possible to arrange the sensor 11 in another part in the anode circuit line 5 or also to arrange it directly in the fuel cell 1. It is also possible to use a temperature sensor that measures ambient temperature. In this way, of course, a switching off in the system moreover containing heat is not of concern.

According to the invention, the freeze protection of the system is possible, so that the concentration C_{MeOH} of the methanol/water mixture is adjusted with respect to the temperature T_{actual} in the anode circuit line 5 and/or with respect to the dominant ambient temperature. The temperature T_{actual} falls as the concentration C_{MeOH} is increased in such a way, and thus the freezing point of the methanol/water mixture is lowered. Thus, the

freeze protection is made possible. With the cold-weather starting of the system, the higher methanol concentration C_{MeOH} allows a faster temperature rise of the fuel cell 1, since methanol is diffused by the membrane 4 to the cathode 3 and is there oxidized immediately after the start of the air supply catalytically under heat. Thus, the cold weather starting procedure is substantially accelerated. Preferably, the temperature monitoring and the associated concentration adaptation take place only upon stopping the system, because in the operation of the fuel cell 1, the temperatures are sufficiently high. However, for other applications, the temperature can also be monitored during the operation.

The sensor 11 continuously monitors the temperature T_{actual} and, if necessary, the concentration C_{MeOH} of the ethanol/water mixture. In the controller 17, the measured temperature T_{actual} is then compared with a given temperature threshold value $T_{threshold}$. Upon stopping the temperature T_{actual} falls under the temperature threshold value $T_{threshold}$, for example, under 0 C, with the methanol concentration C_{MeOH} in the anode circuit line 5 increasing, as additional methanol is tension force-driven into the anode circuit line 5. For this, the injection pump 15 and the fuel injector 14 are triggered accordingly by the controller 17. The concentration increase can take place either via unique addition of a given methanol amount or by exercising control via a concentration monitoring. In the second case, it is possible for the methanol/water mixture to circulate in the anode circuit line 5, at least during the control procedure, with the help of the pump 10 so that the concentration is constantly balanced. Additionally, the concentration sensor 11 is preferably arranged near the fuel injector 14 in the anode circuit line 5, so that during the control with respect to the theoretical value $C_{theo.}$ of the methanol concentration is only then achieved if the concentration over the total anode circuit 5 is extended to the sensor 11.

In the case of control of the methanol concentration C_{MeOH} in the controller 17, the concentration theoretical value $C_{theo.}$ is given as a function of the current temperature T_{actual} and the actual methanol concentration C_{MeOH} is adjusted using ordinary control or monitoring techniques by triggering of the injection pump 15 and the injection valve 14

[sic] to reach the theoretical concentration value $C_{theo.}$. Control can take place, for example, using a characteristic diagram applied in the controller 17, whereby the characteristic diagram shows injection amounts of methanol as a function of the measured temperature T_{actual} and the actual methanol concentration C_{MeOH} in the anode circuit line 5.

Alternatively also several temperature thresholds $T_{threshold\_i}$ can be given, whereby then, if at a decreasing temperature T_{actual}, one falls below the next lower temperature threshold $T_{threshold - i+1}$, in each case a further given methanol amount is used or a higher methanol concentration C_{MeOH} is adjusted. Thus, the system is not always changed over immediately to maximum freeze protection, but the freeze protection is adapted to the actual temperature. Thus, the additional required methanol amount can despite sufficient freeze protection, be reduced and thus the overall efficiency can be improved.

Besides, the fuel cell 1 can be protected in this way if supplemental dangerous components are used in the system containing methanol used at a concentration as a function of the momentary temperature, sufficient for the freeze protection.

Patent Claims

1. Fuel cell system (1) with an anode chamber (2) and a cathode chamber (3), which by a are separated by each other by a proton-conducting membrane (4), with a cathode inlet (8) to supply oxygen-containing gas to cathode chamber (3) and with a cathode exit fuel line (9), anode circuit line (5) to regulate the circuit current activity of a liquid fuel medium/coolant mixture between the anode chamber outlet (6) and the anode chamber inlet (7), along with a device (11) for regulation of the fuel-medium concentration (C_{MeOH}) in anode circuit line (5), a fuel-medium supply vessel (12), a line (13) for the supply of fuel medium from the fuel-medium supply vessel (12) in the anode circuit line (5), with a (13) arranged with device (14) for the dosage of the flowing fuel medium as a function of the fuel medium concentration (C_{MeOH}), characterized by the fact that a device (11) is used for the determination of the temperature (T_{actual}) and that the device (14) is

controllable with respect to the dosage of the to-be-led fuel medium at decreasing temperatures (T_{actual}) in relation to the increase in the fuel-medium concentration (C_{MeOH}) in the anode circuit line (5).

2. Fuel cell system according to Claim 1, characterized by the fact that a device (17) is used for comparing the determined temperature (T_{actual}) with a given temperature threshold value ($T_{threshold}$) and in which the device (14) is controllable with respect to the dosage of the to-be-led fuel medium when falling below the temperature threshold value ($T_{threshold}$) in relation to the increase in the fuel-medium concentration (C_{MeOH}) in the anode circuit line (5).

3. Fuel cell system according to Claim 1, characterized by the fact that a sensor (11) is used for the detection of the ambient temperature (T_{actual}) or the temperature (T_{actual}) of the fuel medium/coolant mixture in the anode circuit line (5).

4. Fuel cell system according to Claim 1, characterized by the fact that a combined concentration and temperature sensor (11) is located in the anode circuit line (5).

5. Fuel cell system according to Claim 3 or 4, characterized by the fact that the sensor (11) between the anode chamber outlet (6) and the metering unit (14) is arranged in the anode circuit line (5).

6. Techniques for driving a fuel cell system with an anode chamber (2) and a cathode chamber (3) which are separated from each other by a proton-conducting membrane (4), whereby cathode chamber (3) with an oxygen-containing gas is acted upon, whereby a liquid fuel medium/coolant mixture is lead with the help of an anode circuit line (5) by the anode chamber (2), and whereby the fuel medium concentration (C_{MeOH}) during the operation of the fuel cell system (1) is held at a given concentration theoretical value ($C_{theo.}$) according to the ambient temperature (T_{actual}) and/or the temperature (T_{actual}) of the

fuel medium/coolant mixture in the anode circuit line (5) and at decreasing temperatures for the monitoring of the performance of sufficient freeze protection with respect to the fuel-medium concentration (C_{MeOH}) increase in the anode circuit line (5).

7. Technique according to Claim 6, characterized by the fact that the theoretical concentration value ($C_{theo.}$) is dependent on the determined temperature T_{actual}.

8. Technique according to Claim 6, characterized by the determined temperature (T_{actual}) constantly being compared with a given temperature threshold value ($T_{threshold}$), and which then, if the determined temperature (T_{actual}) falls below the temperature threshold value (T_{actual}), the fuel-medium concentration (C_{MeOH}) in the anode circuit line (5) is increased.

9. Technique according to Claim 8, characterized by the fact that several temperature thresholds ($T_{threshold\_i}$) are given, whereby then, if at a decreasing temperature (T_{actual}) the next lower temperature threshold ($T_{threshold\_i+1}$) is fallen below, the fuel-medium concentration (C_{MeOH}) in the anode circuit line (5) is increased in each case.

10. Technique according to Claim 8 or 9, characterized by the fact that when falling below one given temperature threshold value ($T_{threshold}$), a given fuel-medium amount into the anode circuit line (5) is admitted or the given theoretical concentration value is increased.

11. Technique according to Claim 6, characterized by the fact that the temperature monitoring is activated only with a switched-off fuel cell system (1).

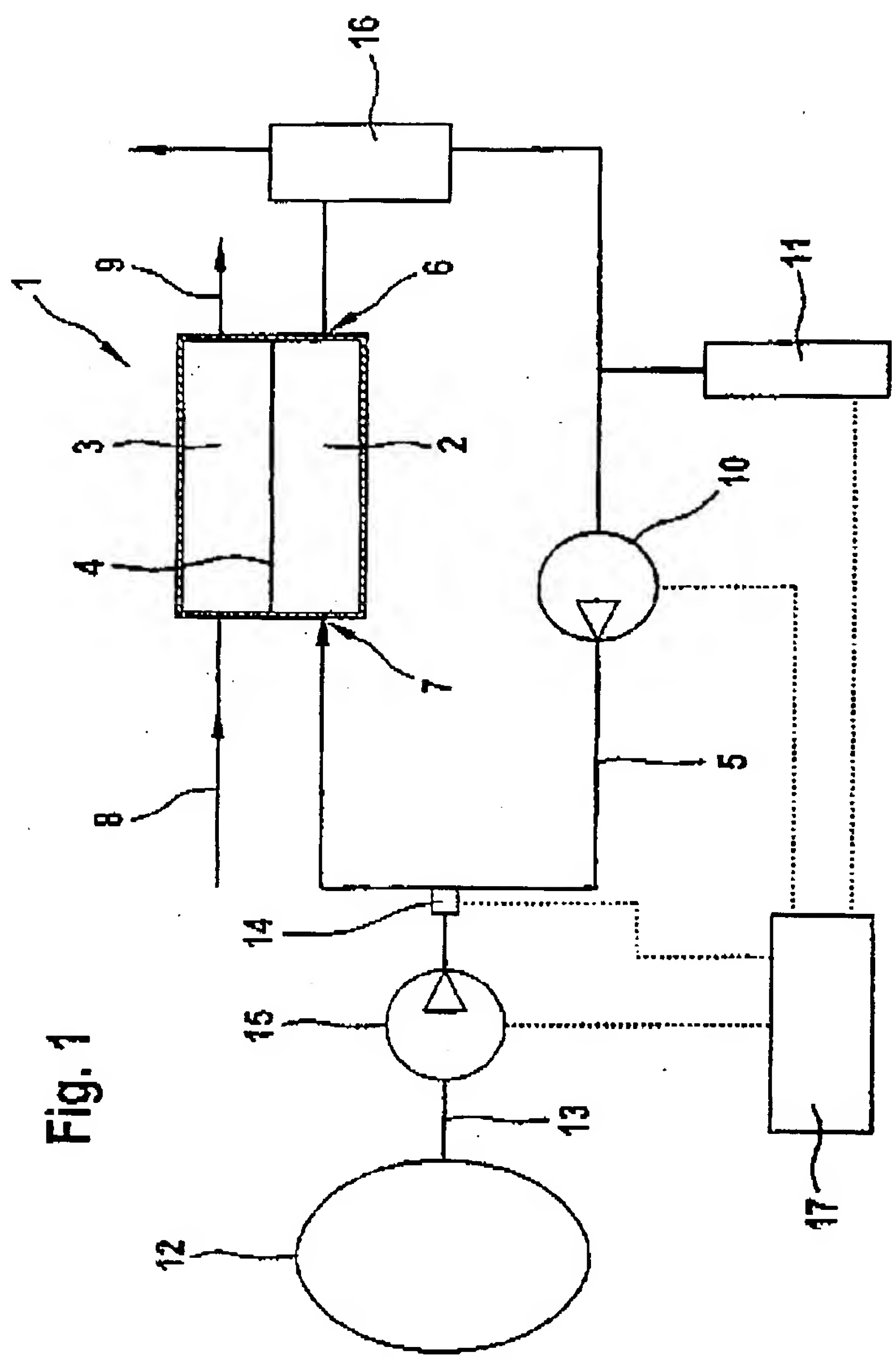


Fig. 1

INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP 00/11587

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H01M8/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No
A	GB 1 304 092 A (ROBERT BOSCH GMBH) 24 January 1973 (1973-01-24) page 2, line 128 -page 3, line 2 claim 1; figure 1 page 3, line 15 - line 20 page 2, line 21 - line 59 ---	1,3,6
A	PATENT ABSTRACTS OF JAPAN vol. 012, no. 461 (E-689), 5 December 1988 (1988-12-05) -& JP 63 184267 A (HITACHI LTD), 29 July 1988 (1988-07-29) abstract ---	1
A	US 5 981 096 A (LAMM ARNOLD ET AL) 9 November 1999 (1999-11-09) column 4, line 52 - line 54 claim 1; figure 1 ---	1

-/--

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
27 April 2001	08/05/2001

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	D'hondt, J

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP 00/11587

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication where appropriate, of the relevant passages	Relevant to claim No.
A	PATENT ABSTRACTS OF JAPAN vol. 010, no. 221 (E-424), 2 August 1986 (1986-08-02) -& JP 61 058170 A (SHIN KOBE ELECTRIC MACH CO LTD), 25 March 1986 (1986-03-25) abstract	1
A	PATENT ABSTRACTS OF JAPAN vol. 1998, no. 13, 30 November 1998 (1998-11-30) -& JP 10 223249 A (TOYOTA MOTOR CORP), 21 August 1998 (1998-08-21) abstract	
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/EP 00/11587

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